Simulation and Computation of the Formation Reactor's Poison: Xe-135 and Sm-149 in Thorium Molten Salt Reactor-500 with MCNP6 Code

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Abstract. Simulation and computation of Thorium Molten Salt Reactor 500 (TMSR-500) have been carried out using the MCNP6 code, referring to the design of MSR-ThorCon by Martingale Inc. USA. Burn up on MSR is run on a high-performance computer. MSR uses fuel as well as the reactor coolant of a liquid mixture of BeF2-NaF-ThF4-UF4. An important information from nuclear reactor operations is the rate of formation of poison nuclides for nuclear reactors, namely Xe-135 and Sm-149, because it has a very high cross section of neutron absorption, respectively 2.65 × 10^6 barn and 4.014 × 10^4 barn. The formation rate of Xe-135 reaches a constant value of 1.65x10^{15} a / cm^3-hour, at the reactor operating time is 50 hours and the k_{eff} is 0.98668. The Sm-149 formation rate reaches a constant value of 1.03x10^{17} a / cm^3-hour at 600 hours operating time with a k_{eff} of 0.96501. When the reactor is shutdown, a Xe-135 peak will be called the xenon dead time, which at this time the reactor should not be turned on because it will cause a neutron buildup. The reactor may be restarted after the Xe-135 concentration equal to its equilibrium, that is after at least 30 hours.

Keywords: Thorium Molten Salt Reactor 500 (TMSR 500), MCNP6, Xe-135, Sm-149.

INTRODUCTION

Thorium Molten Reactor – 500 (TMSR-500) is a reactor designed for the Indonesian market based on MSR-ThroCon by Martingale Inc., USA. Unlike the conventional reactors that use solid fuel, TMSR-500 uses liquid as it fuel. The liquid which is a mixture of liquid salt and nuclear fuel is not only used as a fuel but also used as the reactor's primary coolant [1].

The main reaction that occurs inside the reactors is a controlled chain fission reaction. A fission reaction is a process where a heavy nuclide will split into two lighter nuclides [2]. From the fission reaction, many nuclides called reactor poison accumulated in the core. Reactor poison is a neutron absorber because it has a large cross-section of neutron absorption, so it can cause negative reactivity and decrease in reactor criticality. This can result in a reduction of the fission chain reaction and can cause the reactor to stop operating [3]. The reactor poisons: Xe-135 and Sm-149 can pose a significant threat to the normal operation of the reactor system. So that, research about the Xe-135 and Sm-149 formation is necessary to do. This research aims to detect the Xe-135 and Sm-149 behavior inside the reactor through the simulation method using MCNP6 by modelling the one fuel log geometry to calculate the burn up and the full core geometry to calculate the reactivity.

THEORY

Xe-135 is a poison with the largest neutron absorption cross-section, which is 2.65x10^6 barn [4]. In the reactor, Xe-135 is formed in two ways, form the fission reaction (about 0.3% of the fission result is Xe-135) and from the decay of Te-135 [5]. The equation of reaction from the decay of Te-135 to become a sTABLE nuclide is as follows:

$$^{135}_{52}\mathrm{Te} \xrightarrow[19.0\ \text{second}]{} {}^{135}_{53}\mathrm{I} \xrightarrow[6,6\ \text{hours}]{} {}^{135}_{54}\mathrm{Xe} \xrightarrow[9.1\ \text{hours}]{} {}^{135}_{55}\mathrm{Cs} \xrightarrow[2.3 \times 10^6\ \text{years}]{} {}^{135}_{56}\mathrm{Ba}\ (\text{stable}) \quad (1)$$

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The natural removal of Xe-135 are in two ways, first is the decay of Xe-135 to Cs-135 and the second is through reaction with neutrons like the following equation [4]:

$$54135Xe + 01n \rightarrow 54136Xe + \gamma \quad (2)$$

The concentration of Xe-135 in the reactor will continue to increase until it reaches its equilibrium. The equilibrium of Xe-135 will be achieved when the rate of production of Xe-135 is equal to the rate of removal of Xe-135. The equilibrium Xe-135 concentration can be expressed in the following equation [5]:

$$N_{Xe}(equilibrium) = \frac{(\gamma_{Xe} + \gamma_I)\sum_f^{fuel} \phi}{\lambda_{Xe} + \sigma_a^{Xe}\phi} \quad (3)$$

With:

N_{Xe} = Xe-135 concentration

γ_{Xe} = Xe-135 yield from fission

γ_I = I-135 yield from fission

$\sum_f^{fuel}$ = Macrosopic fission cross-section of fuel

ϕ = Thermal neutron flux

λ_{Xe} = Decay constant of Xe-135

σ_a^{Xe} = Macroscopic absorption cross-section of Xe-135

From Eq. 3, the formation rate of equilibrium Xe-135 can be written as follow:

$$\text{The formation rate of equilibrium Xe-135} = \frac{N_{Xe}}{\Delta t} \quad (4)$$

Δt is the time needed to Xe-135 reaches its equilibrium.

Besides Xe-135, another poison that needs attention is Sm-149 because it has a fairly large absorption cross-section, which is 4.014×10^4. Sm-149 is formed from fission (about 1.08% of fission) and from the decay of Nd-149. The reaction of decaying nuclides to become Sm-149 is as follows:

$$^{149}_{60}\text{Nd} \xrightarrow[1.72\ \text{hours}]{} {}^{149}_{61}\text{Pm} \xrightarrow[51.30\ \text{hours}]{} {}^{149}_{62}\text{Sm (stable)} \quad (5)$$

Sm-149 has different properties from Xe-135, Sm-149 is a sTABLE nuclide, so it can only be removed by absorbing neutrons into Sm-150 as in the following:

$$65149Sm + 01n \rightarrow 65150Sm + \gamma \quad (6)$$

Like Xe-135, the concentration of Sm-149 will continue to increase along the reactor operating time until it reaches its equilibrium. The value of Sm-149 concentration depends on the concentration of Pm-149. The concentration of equilibrium Sm-149 is stated in Equation 7.

$$N_{Sm}(equilibrium) = \frac{\gamma_{Pm}\sum_f^{fuel}}{\sigma_a^{Sm}} \quad (7)$$

With:

N_{Sm} = Sm-149 concentration

γ_{Pm} = Pm-149 yield from fission

$\sum_f^{fuel}$ = Macrosopic fission cross-section of fuel

σ_a^{Sm} = Macroscopic absorption cross-section of Sm-149

From Eq. 7 it can be seen that the concentration of Sm-149 in its equilibrium does not depend on the neutron flux and reactor power, but only depends on the Pm-149 yield and the absorption cross-section of the Sm-149. The rate formation of equilibrium Sm-149 is expressed by Equation 8.

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The formation rate of equilibrium Sm-149 $= \frac{N_{Sm}}{\Delta t}$ (8)

Δt is the time needed to Sm-149 reaches its equilibrium.

To know the concentration of fission products during the reactor's operating period, a burn up calculation must be performed. Riyadi et al (2019) performed burn up calculations using the MCNP6 code for liquid fueled reactors, SAMOP [6]. Burn up is a standard calculation that focuses on the management and control of fuel, including measurement of used fuel, combustion process, processing and the amount of energy produced per unit weight of fuel [7].

The Monte Carlo continuous energy method in the Monte Carlo N-Particle (MCNP) code is one of the reliable methods for transporting particles such as neutrons, photons, and electrons in complex three-dimensional systems. This method is used because it can trace particles from its birth until its death [8].

METHODOLOGY

The procedure of this study is shown in **FIGURE 1.**

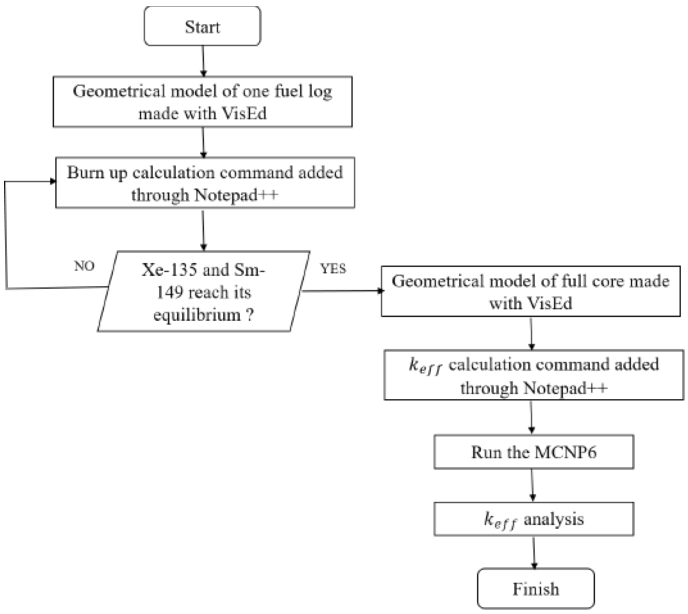

FIGURE 1. Flowchart of the research procedure

The TMSR-500 full core geometry model consists of 84 fuel logs with complex kanal. Burn up calculations with MCNP6 will take longer and larger and more complicated geometry models. To shorten the computational time without reducing the validity of the results can be used periodic boundary condition (PBC) on the fuel log geometry model [9]. The fuel log geometry model is presented in **FIGURE 2.**

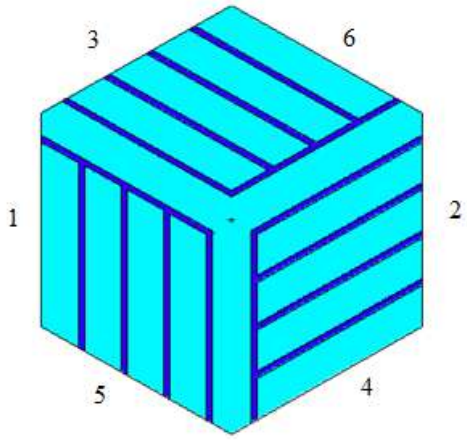

FIGURE 2. One fuel log geometrical model of TMSR-500 [9]

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FIGURE 2 shows the TMSR-500 two dimensional geometry model for burn up calculations made using the Visual Editor (VisEd). Dark blue cells are mixture of BeF2-NaF-UF4-ThF4 with U-235 that enriched to 19.75%, and the light blue cells show graphite moderator.

Side 1 is periodic with side 2 and vice versa. When particles exits the log through the right side (2), it will re-enter the canal through the left side (1). The same is the case with side 3 periodic with side 4 and vice versa, and side 5 periodic with side 6 and vice versa. By applying PBC, the log that is modeled represents one fuel log that is located right in the middle of the core and is surrounded by other logs that repeat infinitely on each side. After the geometry is completed in VisEd, the burn up calculation command is added though the Notepad++, the MCNP6 is run. From the burn up results, it can be known the concentration of Xe-135 and Sm-149, which are then used to calculate the reactor criticality.

The criticality of the reactor is calculated under 2 conditions, at the beginning of the cycle (BOC) when the reactor is clean from poisons and when the reactor poison has been added. The criticality calculation is performed on the full core geometry model as presented in **FIGURE 3**. After the reactor criticality value is obtained, the reactivity calculation is done with the following equation [4]:

$$\rho = \frac{k_{eff} - 1}{k_{eff}} \tag{9}$$

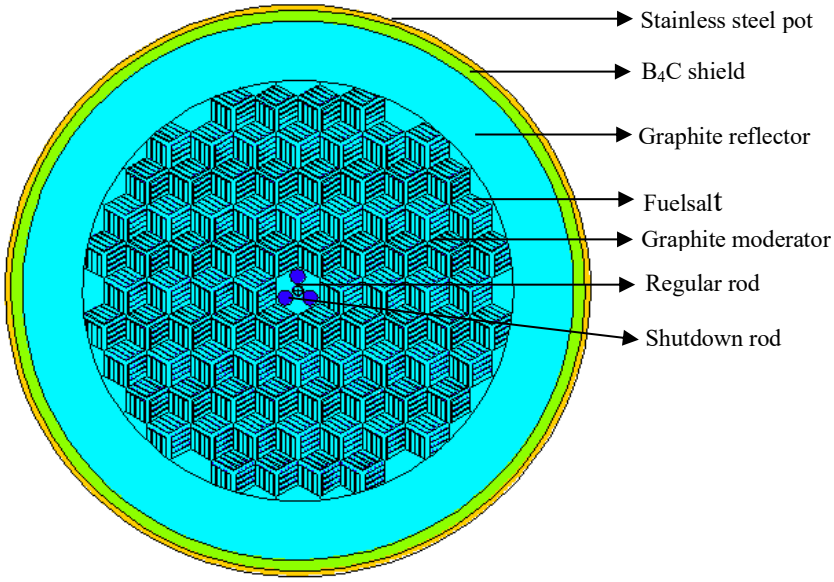

FIGURE 3. Full core geometrical model of TMSR-500 [9]

The simulated burn up time is 600 hours at full power without refueling. When it reaches 600 hours, the sudden shutdown reactor is simulated. A description of the concentrations of Xe-135 and Sm-149 is needed to explain the neutronic characteristics under these conditions. Based on the reactor poison data, ideas will emerge to overcome shutdown conditions and predict the reactor restart processes.

RESULTS AND DISCUSSION

Burn up calculations in MCNP were performed using KCODE with 10^5 simulated particles within 250 cycles and 35 of them being skipped. The only burn up material is fuel. The calculation is done using a high-performance computer and requires 7 days of running.

The k_{eff} value of the one fuel log model is (1.01643 ± 0.00009), and for the full core model, the k_{eff} is (1.01885 ± 0.00008). The difference in the k_{eff} of both models is not too significant, which is 0.238% so that it can be said that the geometry model of the one fuel log correctly represents one log that is located right in the middle of the core. The k_{eff} value in the both geometry model of this study is very close to k_{eff} claim value of TMSR-500 [9] which is 1.00092, with a difference of less than 2%.

FIGURE 4 shows the concentration of Xe-135 against time. At t=0, the concentration of Xe-135 is also 0 which indicates that the reactor is clean and free of reactor poison. At intervals of 0 to 10 hours, the concentration Xe-135 builds up linearly from 0 to 4.74 x 10^{16} ^{a}/cm^3. This build up is caused by the contribution of U-235 fission and Te-135 decay (Te-135 to Xe-135 requires around 6.6. hours). At intervals of 10-30 hours, the concentration

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of Xe-135 shows that the accumulation of Xe-135 has begun to be equal to its removal by neutron absorption to formed Xe-136 and beta decay to formed Cs-135. At 30-50 hours, the concentration of Xe-135 reaches its equilibrium.

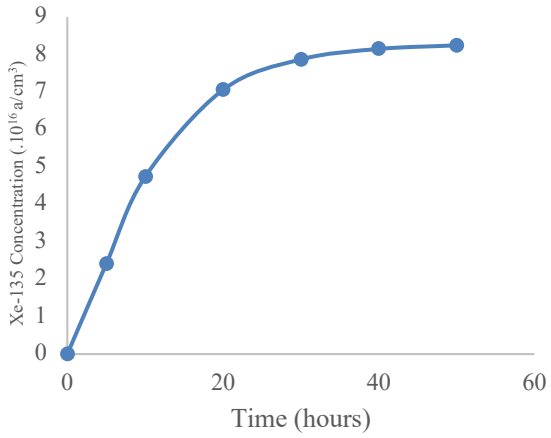

FIGURE 4. Xe-135 concentration at reactor startup

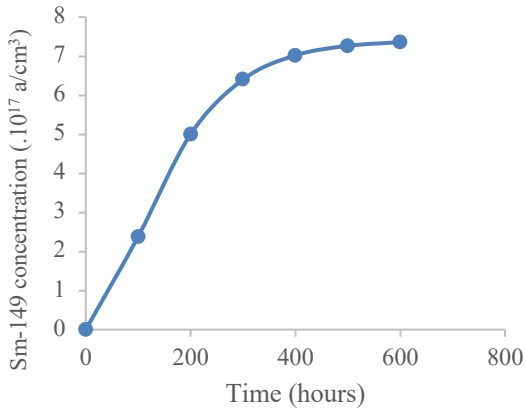

FIGURE 5. Sm-149 concentration at reactor startup

FIGURE 5 shows the Sm-149 concentration against time. At t=0, the Sm-149 concentration also 0, it means that at the beginning of the operation, there's no reactor poison in the core. From 0 to 400 hours of operation, Sm-149 increased from 0 to 7.02 x 10^{17} a/cm^3. From the graph of Sm-149 profile, it can be seen that Sm-149 reaches its equilibrium after 400 hours. Sm-149 takes longer to reach its equilibrium than Xe-135, because aside from the fission product, Sm-149 is also produced by Nd-149 decay that needs about 54 hours to become Sm-149.

FIGURE 6 shows the comparison of Xe-135 and Sm-149 against time at the first 30 days of reactor operation with constant power 250 MW.

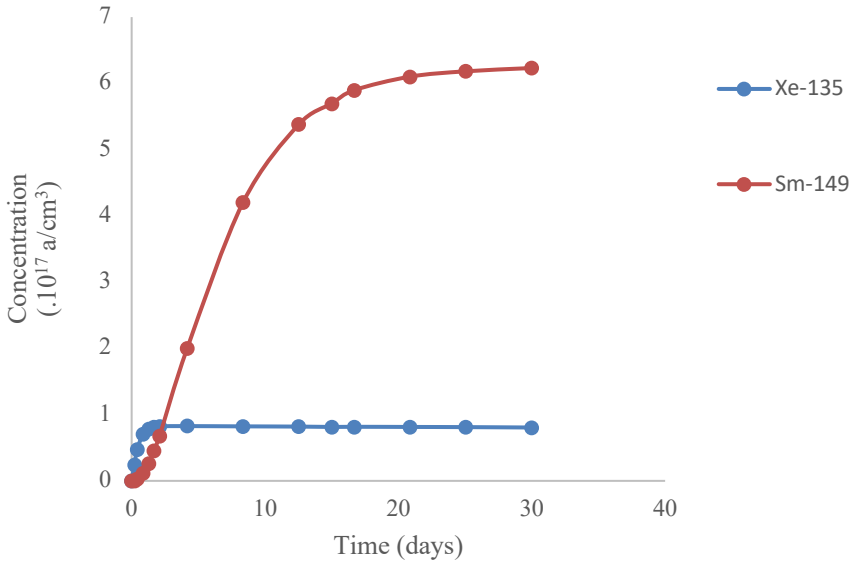

FIGURE 6. Comparison of Xe-135 and Sm-149 at the first 30 days of reactor operation

From **FIGURE 6**, it can be seen that when Xe-135 reaches its equilibrium on the second day of reactor operation, Sm-149 still increasing. Sm-149 reaches its equilibrium after about 20 days of reactor operation. Without the changes in power level, the fission rate of the reactor almost constant for all cycles. As a result, throughout the reactor operation, the concentration of Xe-135 is 1.65x10^{15} a / cm^3-hour and Sm-149 is 1.03x10^{17} a / cm^3-hour. This number is almost the same as the research of Intokiyah and Subhki (2019) that study about reactor poison using GUI [10]. Xenon is a gas poison, so it should be extracted from the core. On TMSR-500, Xe-135 is extracted using spray bubbling methods every 25 minutes with flowing 0.45 grams/hour of helium at the Primary Loop Pump [9].

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The next simulation is a condition when the reactor shutdown suddenly (without minding the cause). **FIGURE 7** and **8** shows the concentration of Xe-135 and Sm-149 when reactor shutdown until restarted.

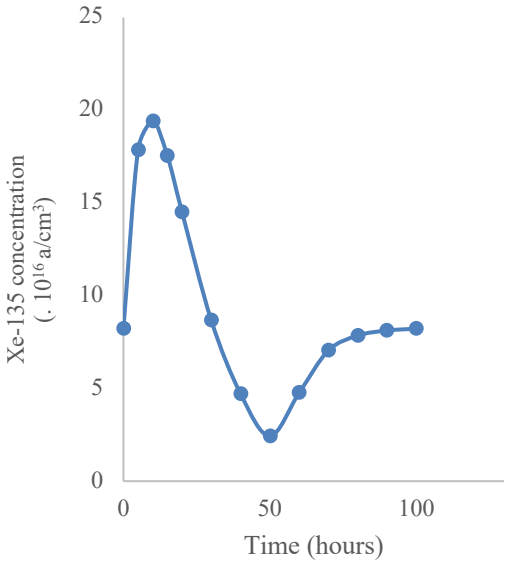

FIGURE 7. Xe-135 concentrations at reactor shutdown and restart

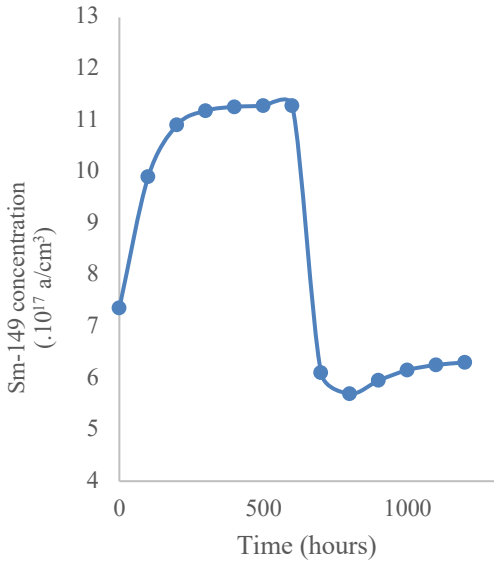

FIGURE 8. Sm-149 concentrations at reactor shutdown and restart

When the reactor is shutdown, the power will drop to 0 MW. This causes the neutron flux in the reactor to drop drastically to zero. Because the neutron flux is zero, there's no Xe-135 formed from fission. However, Xe-135 is still formed from the decay of I-135. **FIGURE 7** shows 10 hours after the shutdown, xenon peak is formed with a concentration of 1.94×10^{17} a/cm^3. The xenon peak is formed because the decay rate of I-135 is faster than the decay rate of Xe-135, and Xe-135 cannot absorb neutrons due to the unavailability of free neutrons. However, the xenon peak does not last long because after a while the amount of Xe-135 formed from the decay of I-135 will decrease and become smaller than the rate of Xe-135 decay so that its concentration will decrease. The height of the xenon peak depends on the magnitude of the neutron flux before the reactor is turned off. The higher the neutron flux value, the higher the peaking.

When the reactor is shutdown, the Sm-149 concentration will not form a peak like Xe-135. The Sm-149 concentration will slowly increase until it reaches a maximum value of 1.13×10^{18} a/cm^3 after the reactor has been turned off for 600 hours. When the reactor power is raised again to 250 MW, the Sm-149 will again absorb neutrons and return to its equilibrium. From **FIGURE 8** we can also know that the reactor will never clean from Sm-149 unless its fuel is renewed because Sm-149 is stable.

The reactivity of Xe-135 and Sm-149 was obtained from the calculation of the difference in reactivity of the reactor when its fuel was clean and when the concentration of Xe-135 or Sm-149 was added. The reactivity values of the two poisons are presented in **TABLE 1**.

TABLE 1. Xe-135 and Sm-149 reactivity

Nuclide		k_{eff}	**Reactivity** ($\%\frac{\Delta k}{k}$)
Fresh fuel		1.01885	1.850
Xe-135	Equilibrium	0.98454	-3.423
	Peak	0.94221	-7.985
Sm-149	Equilibrium	1.01285	-0.584
	Peak	1.00815	-1.044

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Although in FIGURE 6 the concentration of equilibrium Sm-149 is much higher than Xe-135, **TABLE 1** shows that the reactivity value of equilibrium Xe-135 is greater than Sm-149. This is because the Xe-135 absorption cross-section that is 2.65×10^6 barn is larger than the Sm-149 absorption cross-section that is 4.014×10^4 [4], so Xe-135 will absorb more neutrons than Sm-149.

At the peak of Xe-135, the negative reactivity was 57.13% more than the equilibrium Xe-135 reactivity. Positive reactivity is needed which is proportional to the negative reactivity so that the reactor can be operating again. Positive reactivity can be added by lifting the control rod from the core or by adding fissile material. The period time when the reactor cannot return to operation as a result of the Xe-135 influence is called xenon dead time. During the xenon dead time period, positive reactivity should not be added because there can be a large accumulation of neutrons. This is very dangerous because the concentration of Xe-135 continues to decrease until at a certain point the reactor can suddenly start and explode because it cannot handle the accumulation of neutrons that occurs. From **FIGURE 7** it can be seen that the concentration of Xe-135 almost equal to its equilibrium after 30 hours of shutdown. Thus the save minimal time to restarting the reactor is 30 hours.

CONCLUSION

The main reactor poison, Xe-135, reaches equilibrium after 50 hours of reactor operation with a value of 1.65×10^{15} a / cm^3-hour with a reactivity contribution of -3.423 $\% \frac{\Delta k}{k}$. The second poison, Sm-149 reached the equilibrium after 600 hours of reactor operation with a value of 1.03×10^{17} a / cm^3-hour with a reactivity contribution of -0.584 $\% \frac{\Delta k}{k}$. After reaching equilibrium, the concentration of Xe-135 and Sm-149 will tend to be constant if there is no change in reactor power. When the reactor is shutdown, a Xe-135 peak will be called the xenon dead time, which at this time the reactor should not be turned on because it will cause a neutron buildup. The reactor may be restarted after the Xe-135 concentration equal to its equilibrium, that is after at least 30 hours.

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